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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of regional background particulate matter concentration based on vertical distribution characteristics" *by* S. Han et al.

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Comment: General This is a commendable exercise in interpretation of tall tower aerosol results. In the introduction an overview over tall tower aerosol data interpretation(e.g.,Brown et al.,2013;Heintzenberg et al.,2008; Andreae et al.,2015) should put the present approach into perspective. The main weakness is a lack of quantification of the scales that are derived from the study. Response: The overview over tall tower aerosol data interpretation(e.g.,Brown et al.,2013;Heintzenberg et al.,2008; Andreae et al.,2015) has been added in the introduction. Measurements at different heights within the boundary layer could represent different horizontal scales of pollution. Ac-

cording to our study, the nocturnal PM2.5 mass concentration time series with the 6-10 days period at the height of 220m can reflect the influence of regional pollution within 102 km away from the measurement tower. The regional scale in this study has been quantified in the revised manuscript.

Language The English still needs substantial improvements. Examples: transform of PM2.5, associated with each other among cities, vertical height, surface layer is closely related, change rules, variation rules of temperature, were in effect Response: The revised manuscript has been edited by a master of the English language.

Recommendation Accept after revision according to comments

Detailed comment: Comment: Page 14891, Line 1. What is " regional compound pollution" Response: It has been revised as "air pollution complex" (Shao et al., 2006) in the manuscript. The air pollution complex is characterized by an increase in the oxidizing capacity of the atmosphere, reduced atmospheric visibility, and the deterioration of environmental quality throughout the entire region; It features the interactions between the sources and sinks of air pollutants, the coupling processes of the transformation of pollutants, and the synergetic environmental impacts of air pollutants (Zhu et al., 2011).

Shao M, Tang X, Zhang Y, et al. City clusters in China: air and surface water pollution[J]. Frontiers in Ecology and the Environment, 2006, 4(7): 353-361. Zhu T, Shang J, Zhao D F. The roles of heterogeneous chemical processes in the formation of an air pollution complex and gray haze[J]. Science China Chemistry, 2011, 54(1): 145-153.

Comment: Page 14891, Line 4. "Secondary chemical reactions" have not been introduced by Chinese scientists. Refer to appropriate textbooks instead. Response: In the revised manuscript, it has been modified as follows. "The origin of PM is complex. It involves both primary emissions as well as secondary particle production due to chemical reactions in the atmosphere".

Comment: Page 14892, Line 8. Particle size distribution should be listed under "Phys-

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ical method" Response: Particle size distribution has been listed under the "physical method" in the revised manuscript.

Comment: Page 14892, Line 22. There are established concepts in atmospheric dynamics that could be applied here more specifically such as footprints (e.g. Schmid,2002; Foken, 2008). Response: To interpret the spatial representativeness of vertical measurement, the footprint concept has been added in the revised manuscript. The footprint concept is capable of linking observed data to spatial context. The integral beneath the foot-print function expresses the total surface inïňĆuence on the signal measured by the sensor at height above the surface (Schmid, 2002; Ding et al., 2005; Foken, 2008). Three main factors affecting the size and shape of flux footprint are: measurement height, surface roughness, and atmospheric stability. Increase in measurement height, decrease in surface roughness, and change in atmospheric stability from unstable to stable would lead to an increase in size of the footprint and move peak contribution away from the instrument (https://en.wikipedia.org/wiki/Flux_footprint).

Schmid H P. Footprint modeling for vegetation atmosphere exchange studies: a review and perspective [J]. Agricultural and Forest Meteorology, 2002, 113(1): 159-183. Ding G, Chen Z, Gao Z, et al. The vertical structure and its dynamic characteristics of PM10 and PM2. 5 in lower atmosphere in Beijing city [J]. Science in China, Series D, 2005, 35(S1): 31-44. Foken T, Nappo C J. Micrometeorology[M]. Springer Science & Business Media, 2008.

Comment: Page 14893, Line 24. More specific information about PM2.5 measurements are requested: Total time period, temporal resolution, uncertainties. Response: Mass concentrations of PM2.5 were measured using ambient particulate monitor chemiluminescence (TEOMR-RP1400a) at four levels (2m,40m,120m, and 220m) from July 1 to September 30,2009. The monitor's data output consists of 1-hour and 24-hour average mass concentration updated every 10 minutes and on the hour ,with the precision of $\pm 1.5 \mu$g/m3(1-hour ave) and $\pm 0.5 \mu$g/m3(24-hour ave) respectively. Accuracy for mass measurement is $\pm 0.75\%$.

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Comment: Page 14894, Line 3. More information is requested on the PM10 sampling: PM10 inlet characteristics (as function of wind speed), start/stop of the 24 h samples. Response: Twenty-four hour PM10 samples were collected from local Beijing time 08:00 to 07:00 the next day using medium-volume PM10 samplers (TH-150,Wuhan Tianhong Intelligence Instrumentation Facility) at the heights of 10 m, 40 m, 120 m, and 220 m from August 24 to September 12, 2009. The sampler has a system of automatic constant-flow control. Flow rate of sampling in this study is 100 L min-1, and the relative error of flow is less than 3%.

Comment: Page 14894, Line 20.Definition of "seasons" Response: The four seasons were designated as March to May for spring, June-August for summer, September-November for autumn, and December-February the next year for winter.

Comment: Fig 2. Typo in legend Response: The typo error "Netural" has been corrected to "Neutral".

Comment: Page 14894, Line 15. Uncertainties of chemical analyses are missing Response: Filters were conditioned for 48 h in darkened desiccators before and after sampling prior to gravimetric determination. The filters were weighed on a electronic microbalance(AX205,Mettler-Toledo, LLC) with a $\pm$0.01mg sensitivity in a clean room under constant temperature(20$\pm$1âĎČ) and RH(40$\pm$3%). Samples were stored air-tight in a refrigerator at about 4âĎČ before chemical analyses. Elements(Si,Ti,Al,Mn,Ca,Mg,Na,K,Cu,Zn,Pb,Cr,Ni,Co,Fe and V) were analyzed by Inductively Coupled Plasma-atomic emission spectroscopy(ICP 9000(N+M)Thermo Electron Corporation, USA). Blank filters were processed simultaneously with sample filters. Ultrapure water, both unfiltered and filtered, and nitric acid were also analyzed. The average element values in the blanks were subtracted from those obtained for each sample filter. 10 percent of total samples were analyzed in duplicate to verify sample homogeneity. The precision and accuracy were checked by analysis of an intermediate calibration solution. Extraction efficiencies were evaluated by analysis of the certified reference material from National Research Center of CRM. The recovery

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value was between 85% and 110%. A calibration check was performed to ensure a relative error no more than 2% for major elements and 5% for trace elements. Water-soluble ions(NH4+,Cl-, NO3- and SO42-) were analyzed by ion chromatography (DX-120, Dionex Ltd., USA) after extraction by deionized water. External calibration was employed to quantify the ions concentrations. A calibration check with external standards was performed to ensure a relative error no more than 10%. The uncertainty contributions of the calibration curve, calibration solution, and repetitive measurement for unknown sample were taken into account. The expanded uncertainty was 3.8% with a coverage factor k=2. The thermal optical carbon analyzer (Desert Research Institute (DRI) Model 2001, Atmoslytic Inc., Calabasas, CA, USA) was used to measure organic carbon (OC) and elemental carbon (EC). The heating process can be found in IMPROVE_A protocol (Chow et al., 2010,2011; Cao et al.2003). Field blank and lab blank were considered and all sampling concentrations were revised by blank concentration. The uncertainty contributions of the calibration curve, calibration solution, and repetitive measurement for unknown sample were taken into account. The expanded uncertainty was 7.6% with a coverage factor k=2.

Chow J C, Watson J G, Chen L W A, et al. Quantification of PM 2.5 organic carbon sampling artifacts in US networks [J]. Atmospheric Chemistry and Physics, 2010, 10(12): 5223-5239. Chow J C, Watson J G, Robles J, et al. Quality assurance and quality control for thermal/optical analysis of aerosol samples for organic and elemental carbon [J]. Analytical and bioanalytical chemistry, 2011, 401(10): 3141-3152. Cao J J, Lee S C, Ho K F, et al. Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period[J]. Atmospheric Environment, 2003, 37(11): 1451-1460.

Comment: Page 14894, Line 24.Why should a temperature profile “correlate” with height? Response: In the revised manuscript, it has been modified as follows. The vertical profile of wind speed and temperature under different stability are shown in Fig 2. In low atmosphere, weak vertical gradient in the temperature profile was observed

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over 100m. Similarly, small vertical gradient in wind speed was found over 150m.

Comment: Page 14895, Line 12.Details on hourly PM10 measurements are missing. Response: The expression is not clear. It has been modified as follows. In this study, hourly averaged PM2.5 concentration measurement and twenty-four hour PM10 sampling were conducted at four platforms (10, 40, 120, and 220m). Details on PM10 sampling, as is stated above, have been added in the revised manuscript (section 2.2).

Comment: Page 14895, Line 22. Is there no seasonal variation in turbulent intensity? Response: Diurnal variations of three dimensional components of turbulent intensity at 3 different heights in each season are shown in the Figure below (Supplemental Fig. S1 in the revised manuscript). As a whole, the averaged diurnal variations of turbulent intensity in each season were reflecting the same trends. The diurnal peaks appeared later and turbulent intensity was slightly weaker in winter than in other seasons.

Supplemental Fig. S1 Diurnal variations of three dimensional components of turbulent intensity at 3 different heights in each season

Comment: Fig 5. Why are only fall data discussed and shown in Fig.5? Response: Fig.5 shows the vertical diurnal variation of PM2.5 mass concentrations during the period from July 1 to September 30, 2009. The four seasons were designated as March to May for spring, June-August for summer, September-November for fall, and December-February the next year for winter. Therefore part of the summer and fall data were discussed and shown in Fig.5. The title of Fig.5 has been corrected in the revised manuscript.

Comment: Page 14896, Line 8.How can aerosol particles emitted near the ground "accumulate" at 120m during the night? Response: The expression is ambiguous. In the revised manuscript, it has been modified as follows. The nocturnal planetary boundary layer(NPBL) height generally ranges from 100 m to 150 m(Fig. 3). At the 1st and 2nd platform (2m, 40 m), the measured PM are normally at inside of the NPBL. By contrast, the measurement platform at 220 m is generally outside the NPBL. The

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level 3 (120 m) is considered as at the transition zone between inside and outside of the NPBL. Due to the dynamical stability of the NPBL, the vertical mixing of pollutants between inside and outside of the NPBL is very weak. The surface emitted PM are normally trapped inside the NPBL, leading to the difference in the amount of aerosols below and above the NPBL. Among these 4 platforms (2 m, 40 m, 120 m and 220 m), PM2.5 concentration at 220m during the night is the lowest. This indicates that the observation value of 220 m at night is less affected by local sources of emission and is largely attributed to regional scale pollution.

Comment: Page 14896, Line 14. It should be possible to calculate the PBL height throughout the day and relate the vertical particle profiles to that height throughout the day. Response: Impact of the PBL on the vertical particle profiles has been illustrated in the revised manuscript. The vertical variation patterns of PM2.5 concentrations were quite different during the daytime and night resulting from a combination of diurnal variations of emissions and planetary boundary layer (PBL). After sunrise, the PBL starts to rapidly increase, pollutants near the ground gradually diffuse upward and the PM2.5 concentration near the surface gradually decreases. At noontime, the mixing layer is fully developed with the averaged PBL height being about 1000-1200m. Among these 4 platforms (2 m, 40 m, 120 m and 220 m), PM2.5 concentration at 220m is the highest during noon-afternoon-time. In contrast, after 6 PM, the PBL starts to rapidly decrease. The nocturnal planetary boundary layer(NPBL) height generally ranges from 100 m to 150 m. At the 1st and 2nd platform (2 m, 40 m), the measured PM are normally at inside of the NPBL. By contrast, the measurement platform at 220 m is generally outside the NPBL. The level 3 (120 m) is considered as at the transition zone between inside and outside of the NPBL. Due to the dynamical stability of the NPBL, the vertical mixing of pollutants between inside and outside of the NPBL is very weak. The surface emitted PM are normally trapped inside the NPBL, leading to the difference in the amount of aerosols below and above the NPBL. Among these 4 platforms, PM2.5 concentration at 220m at night is the lowest. This indicates that the observation value of 220 m at night is less affected by local sources of emission and is

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largely attributed to regional scale pollution.

Comment: Page 14896, Line 18.The reader still does not know where the PM10 data come from, are they the sum of all analyzed chemical components. Response: Twenty-four hour PM10 samples were collected from local Beijing time 08:00 to 07:00 the next day using medium-volume PM10 samplers (TH-150,Wuhan Tianhong Intelligence Instrumentation Facility) at the heights of 10 m, 40 m, 120 m, and 220 m from August 24 to September 12, 2009. More information on the PM10 sampling and chemical analyses has been illustrated in the above responses and been added in the section 2.2 in the revised manuscript. Please see the above-mentioned response to the comments(Page 14894, Line 3; Page 14894, Line 15; Page 14895, Line 12).

Comment: Page 14897, Line 5. Of what use are the coefficients of divergence? Response: Coefficients of divergence (CD) analysis has been used to assess spatial variability. The CD values provide information on the degree of uniformity between sampling sites (Krudysz et al., 2009). In this study, CD analysis was used to assess vertical variability of chemical elements in PM10 samples collected at 4 heights.

Krudysz M, Moore K, Geller M, et al. Intra-community spatial variability of particulate matter size distributions in Southern California/Los Angeles[J]. Atmospheric Chemistry and Physics, 2009, 9(3): 1061-1075.

Comment: Page 14897, Line 25. Due to potentially high chlorine losses Cl as marker for sea salt is rather uncertain(Klockow et al.,1979). Response: The chlorine loss definitely is a common phenomenon in the gas chemical processes of the sea salt. Many Cl- in sea salt could be replaced by SO42- or NO3- and released in HCl. In this study, we consider Cl- as the marker of sea salt mainly because sea salt is the dominant source of Cl- in our research region. Even after the potentially high chlorine losses, we still found certain amount of Cl- in the PM. The purpose of the marker in this study is not to quantity how many the sea salt is, but only to get an insight into the qualitative result of the sea salt. We totally agree with the comment that such a marker

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may cause some uncertainty, so no definite conclusion on the sea salt's contribution was drawn in our manuscript.

Comment: Page 14898, Line 17.Are all seasons combined in the CMB modeling? Response: Not all seasons were combined in the CMB modeling. Ambient PM10 sampling in this study was conducted from August 24 to September 12, 2009. The dataset of chemical composition in the PM10 samples during the measurement period were used in the CMB modeling.

Comment: Fig. 7. The filtering results do not look convincing in comparison to the unfiltered data. The wild swings in the filtered data need to be justified and need to be explained in terms of underlying meteorological processes. Response: The wild swings in the PM2.5 concentration data were mainly resulted from several different meteorological processes during the measurement. The data used in this study were collected at a 255-m meteorological tower which is located at the atmospheric boundary layer observation station (WMO Id.No. 54517, 39o04'29.4"N, 117o12'20.1"E) in Tianjin. According to the meteorological dataset of that station, precipitation processes were recorded during the period of 22-24 July, with the amounts of rainfall ranged from 3.2 to 94.6mm, followed by a rapid decrease in PM2.5 concentration on 25 July due to consequent cleaning of the air. Then, beginning on 26 July, mist paired with calm winds caused a build-up of PM2.5 concentration until July 29. Similar meteorological processes were reported during the period of 22-25 of August, 4-9 and 20-25 of September, which resulted in the cycle of cleaning and build-up of air pollutants.

Comment: Page 14903, Line 3. Do not report more significant figures in concentration than what corresponds to the uncertainty of the data, here certainly no more than 2 significant figures. Response: In the revised manuscript, it has been modified as follows. The averaged regional background PM2.5 concentrations in July, August and September, 2009 were 40 $\pm$20μg/m3ïijŇ64 $\pm$17 μg/m3 and 53$\pm$11 μg/m3, respectively.

Comment: Page 14903, Line 13. Explain what you mean with "special stratification" in

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terms of standard boundary layer meteorology. Response: The atmospheric layer at 100-150m is considered as a special stratification, the variation patterns of temperature and wind speed with height were different compared with the upper and lower layers. The vertical profile of wind speed and temperature under different stability are shown in Fig 2. In low atmosphere, weak vertical gradient in the temperature profile was observed over 100m. Similarly, small vertical gradient in wind speed was found over 150m. Besides, from the height of 40 m to 120 m, the u, v and w components of turbulent intensity reduced by 27%, 32% and 21%, respectively. From 120 m to 220 m, the u, v and w components reduced by 12%, 13% and 15%, respectively. The descending trend is more obvious from 40 m to 120 m than that of from 120 m to 220 m. This indicates that there were fully vertical and horizontal turbulence exchanges below 120m of the tower, but relatively weaker exchanges over 120m.

Comment: Page 14903, Line 20. Here and previously in the text the term regional scale needs to be quantified. Response: Measurements at different heights within the boundary layer could represent different horizontal scales of pollution. According to our study, the nocturnal PM2.5 mass concentration time series with the 6-10 days period at the height of 220m can reflect the influence of regional pollution within 102 km away from the measurement tower. That is to say, regional scale is about 102 km in this study.

Comment: Page 14904, Line 20. What do you mean by “better”? (also on page 14902 26) Response: The purpose of this study is to characterize the regional pollution contribution and to evaluate regional background PM concentration levels. However, regional background concentration can hardly be measured directly. Original PM concentration time series measured on the ground reflect a combination of influence from local pollution and regional-scale pollution. A method to estimate regional background PM concentration is proposed in this paper, based on the vertical variation periodic characteristics of the atmospheric boundary layer structure and particle mass concentration, as well as the vertical distribution of chemical composition and

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pollution source apportionment. The measurement height influenced relatively less by local pollution emission was determined and impacts from local-scale pollution on the short-term fluctuations have been removed from the original PM concentration by wavelet transformation. After the filtering, regional-scale pollution was "better" represented in the remaining part compared with the original PM concentration time series. More explaination has been added in the revised manuscript.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C6418/2015/acpd-15-C6418-2015-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 14889, 2015.